alkylbenzene

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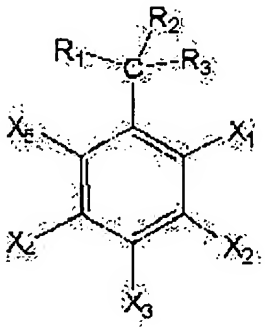
(54) NONAQUEOUS ELECTROLYTE AND LITHIUM SECONDARY BATTERY USING THE SAME

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a lithium secondary battery excellent in battery properties such as cycle property, electric capacity and a shelf life of the battery.

SOLUTION: This nonaqueous electrolyte with an electrolyte dissolved in a nonaqueous solvent contains 0.1-20 wt.% of a tert-alkylbenzene derivative expressed by general formula (I), wherein R1 is a 2-4C alkyl group; R2 and R3 are each a 1-4C alkyl group; and X1, X2, X3, X4 and X5 are independently a hydrogen atom, a 1-12C hydrocarbon group or a halogen atom. The lithium secondary battery using the nonaqueous electrolyte is provided.



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* NOTICES *

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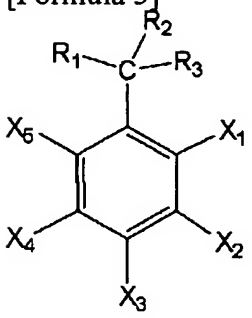
3.In the drawings, any words are not translated.

DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention] This invention relates to the nonaqueous electrolyte which can offer the lithium secondary battery excellent also in cell properties, such as the cycle property of a cell, and electric capacity, a preservation property, and the lithium secondary battery using it.
[0002]
[Description of the Prior Art] In recent years, the lithium secondary battery is widely used as power sources for a drive, such as small electronic equipment. The lithium secondary battery mainly consists of a positive electrode, nonaqueous electrolyte, and a negative electrode, and the lithium secondary battery which used lithium multiple oxides, such as LiCoO2, as the positive electrode, and used the carbon material or the lithium metal as the negative electrode is used especially suitably. And as nonaqueous electrolyte for the lithium secondary batteries, carbonate, such as ethylene carbonate (EC) and propylene carbonate (PC), is used suitably.
[0003]
[Problem(s) to be Solved by the Invention] However, the rechargeable battery which has the further excellent property about cell properties, such as the cycle property of a cell and electric capacity, is called for. When the solvent in nonaqueous electrolyte carries out oxidative degradation of the part locally at the time of charge, the lithium secondary battery using LiCoO2, LiMn2O4, LiNiO2, etc. as a positive electrode produces cell performance degradation, in order that this decomposition product may check the desirable electrochemical reaction of a cell. This is considered to originate in the electrochemical oxidation of the solvent in the interface of a positive-electrode ingredient and nonaqueous electrolyte. Moreover, while the lithium secondary battery using high-crystalized carbon materials, such as a natural graphite and an artificial graphite, repeats charge and discharge also in EC for which the solvent in nonaqueous electrolyte carries out reduction decomposition on a negative-electrode front face at the time of charge, and is generally widely used as a nonaqueous electrolyte solvent at it as a negative electrode, reduction decomposition takes place in part, and cell performance degradation happens. For this reason, the present condition is that cell properties, such as the cycle property of a cell and electric capacity, are not necessarily satisfactory.
[0004] This invention aims at offering the nonaqueous electrolyte for lithium secondary batteries which can constitute the lithium secondary battery which solved the technical problem about the above nonaqueous electrolyte for lithium secondary batteries, was excellent in the cycle property of a cell, and was further excellent also in cell properties, such as electric capacity and a preservation property in a charge condition, and the lithium secondary battery using it.
[0005]
[Means for Solving the Problem] It sets to the nonaqueous electrolyte by which the electrolyte is dissolved in the non-aqueous solvent, and this invention is the following general formula (I) in this nonaqueous electrolyte.
[0006]

[Formula 3]



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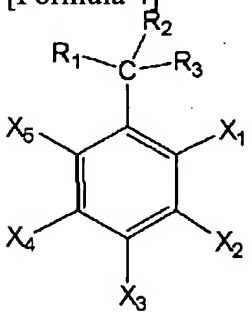
[0007] (-- R1 shows the alkyl group of carbon numbers 2-4 among a formula, R2 and R3 show independently the alkyl group of carbon numbers 1-4, respectively, and X1, X2, X3, X4, and X5 are the hydrocarbon groups or halogen atoms of a hydrogen atom and carbon numbers 1-12 independently, respectively.) -- it is related with the nonaqueous electrolyte characterized by the tert-alkylbenzene derivative expressed containing 0.1 to 20% of the weight. Moreover, it sets to the lithium secondary battery which consists of nonaqueous electrolyte by which the electrolyte is dissolved in the positive electrode, the negative electrode, and the non-aqueous solvent, and this invention is the following type (I) in this nonaqueous electrolyte.

[0008]

[Formula 4]



(I)

[0009] (-- R1 shows the alkyl group of carbon numbers 2-4 among a formula, R2 and R3 show independently the alkyl group of carbon numbers 1-4, respectively, and X1, X2, X3, X4, and X5 are the hydrocarbon groups or halogen atoms of a hydrogen atom and carbon numbers 1-12 independently, respectively.) -- it is related with the lithium secondary battery characterized by the tert-alkylbenzene derivative expressed containing 0.1 to 20% of the weight.

[0010] The nonaqueous electrolyte of this invention is used as a configuration member of a lithium secondary battery. Especially about configuration members other than the nonaqueous electrolyte which constitutes a rechargeable battery, it is not limited but the various configuration members currently used conventionally can be used.

[0011]

[Embodiment of the Invention] In the tert-alkylbenzene derivative expressed with said general formula (I) contained in the nonaqueous electrolyte by which the electrolyte is dissolved in the non-aqueous solvent, R1 shows the alkyl group of carbon numbers 2-4 like an ethyl group, a propyl group, and butyl, R2 and R3 become independent, respectively, and its alkyl group of carbon numbers 1-4 like a methyl group, an ethyl group, a propyl group, and butyl is desirable. At this time, straight chain-like an alkyl group or a branching-like alkyl group is sufficient as an alkyl group. Moreover, X1, X2, X3, X4, and X5 become independent, respectively, and its alkyl group of the shape of branching, such as an alkyl group of the shape of a straight chain, such as a hydrogen atom, a methyl group, an ethyl group, a propyl group, and butyl, and an iso-propyl group, iso-butyl, sec-butyl, tert-butyl, a tert-pentyl radical, is desirable. Moreover, you may be the cycloalkyl radical of the carbon numbers 3-6, such as a cyclo

propyl group and a cyclohexyl radical. Furthermore, you may be phenyl groups by which alkylation was carried out, such as a tolyl group besides a phenyl group and benzyl, a tert-buthylphenyl radical, tert-butyl benzyl, and a tert-pentyl phenyl group, and benzyl. Furthermore, a halogen atom like a fluorine atom, a chlorine atom, a bromine atom, or an iodine atom is desirable again. It is desirable to have the hydrocarbon group or halogen atom of such carbon numbers 1-12.

[0012] As an example of a tert-alkylbenzene derivative expressed with said general formula (I) For example, a tert-pentyl benzene [R1= ethyl group, an R2=R3= methyl group, X1=X2=X3=X4=X5= hydrogen atom], a benzene (1-ethyl-1-methylpropyl) [R1=R2= ethyl group, An R3= methyl group, X1=X2=X3=X4=X5= hydrogen atom], and benzene (1 and 1-diethyl propyl) [R1=R2=R3= ethyl group, X1=X2=X3=X4=X5= hydrogen atom], a benzene (1 and 1-dimethyl butyl) [R1= propyl group, An R2= methyl group, an R3= methyl group, X1=X2=X3=X4=X5= hydrogen atom], A benzene [R1= propyl group, an R2= ethyl group, (1-ethyl-1-methylbutyl) An R3= methyl group, X1=X2=X3=X4=X5= hydrogen atom], a benzene (1-ethyl-1-ethyl butyl) [R1= propyl group, An R2= ethyl group, an R3= ethyl group, X1=X2=X3=X4=X5= hydrogen atom], benzene (1, 1, 2-trimethyl propyl) [an R1=iso-propyl group, an R2= methyl group, an R3= methyl group, and a X1=X2=X3=X4=X5= hydrogen atom], etc. are mentioned. As an example of a tert-pentyl benzene derivative, moreover, a 1-methyl-4-tert-pentyl benzene [R1= ethyl group, R2=R3= methyl group, X1=X2=X4=X5= hydrogen atom, and X3= methyl group], 5-tert-pentyl-meta xylene, a [R1= ethyl group, an R2=R3= methyl group, A X1=X3=X5= hydrogen atom and X2=X4= methyl group], 1, and 3-G tert-pentyl benzene [R1= ethyl group, R2=R3= methyl group, X1=X3=X4=X5= hydrogen atom, and X2=tert-pentyl radical], A 1 and 4-G tert-pentyl benzene [R1= ethyl group, an R2=R3= methyl group, X1=X2=X4=X5= hydrogen atom and X3=tert-pentyl radical], 1, 3, a 5-tree tert-pentyl benzene [R1= ethyl group, R2=R3= methyl group and X1=X3=X5= hydrogen atom and X2=X4=tert-pentyl radical], A 4-BUROMO-tert-pentyl benzene [R1= ethyl group, an R2=R3= methyl group, X1=X2=X4=X5= hydrogen atom and X3= bromine], a 4-fluoro-tert-pentyl benzene [R1= ethyl group, R2=R3= methyl group, X1=X2=X4=X5= hydrogen atom, and X3= fluorine], A 4-chloro-tert-pentyl benzene [R1= ethyl group, an R2=R3= methyl group, X1=X2=X4=X5= hydrogen atom and X3= chlorine], 4-iodine-tert-pentyl benzene [an R1= ethyl group, an R2=R3= methyl group, a X1=X2=X4=X5= hydrogen atom, and X3= iodine], etc. are mentioned.

[0013] If there are too many contents of the tert-alkylbenzene derivative expressed with said formula (I) contained in nonaqueous electrolyte, sufficient cell engine performance which the cell engine performance may fall and was expected to be too few will not be obtained. Therefore, since the cycle property of range [0.5 - 5% of the weight of] improves preferably especially 0.2 to 10% of the weight 0.1 to 20% of the weight to the weight of nonaqueous electrolyte, the content is good.

[0014] As a non-aqueous solvent used by this invention, for example Ethylene carbonate (EC), Propylene carbonate (PC), butylene carbonate (BC), Annular carbonate, such as vinylene carbonate (VC), and lactone, such as gamma-butyrolactone Dimethyl carbonate (DMC), methylethyl carbonate (MEC), Chain-like carbonate, such as diethyl carbonate (DEC), a tetrahydrofuran, 2-methyl tetrahydrofuran, 1,4-dioxane, 1, 2-dimethoxyethane, Amides, such as ester, such as nitril, such as ether, such as 1, 2-diethoxy ethane, 1, and 2-dibutoxy ethane, and an acetonitrile, methyl propionate, pivalate methyl, and pivalate octyl, and dimethylformamide, are mentioned.

[0015] These non-aqueous solvents may be used by one kind, and may be used combining two or more kinds. Although especially the combination of a non-aqueous solvent is not limited, various combination, such as combination of annular carbonate and chain-like carbonate, combination of annular carbonate and lactone, and combination of three kinds of annular carbonate and chain-like carbonate, is mentioned, for example.

[0016] As an electrolyte used by this invention, for example LiPF6, LiBF4, LiClO4, LiN (SO2CF3) 2, LiN (SO2C2F5)2 and LiC (SO2CF3)3, LiPF4(CF3) 2, LiPF3(C2F5) 3, LiPF3(CF3) 3, LiPF3(iso-C3F7) 3, LiPF5 (iso-C3F7), etc. are mentioned. These electrolytes may be used by one kind, and they may be used, combining them two or more kinds. 0.1-3 M of these electrolytes is usually preferably used by the concentration of 0.5-1.5M, dissolving in the aforementioned non-aqueous solvent.

[0017] The electrolytic solution of this invention is obtained by mixing the aforementioned non-aqueous

solvent, dissolving the aforementioned electrolyte in this, and dissolving at least one sort in the tert-alkylbenzene derivative expressed with said formula (I).
[0018] The electrolytic solution of this invention is suitably used as the configuration member of a rechargeable battery, especially a configuration member of a lithium secondary battery. Especially about configuration members other than the electrolytic solution which constitutes a rechargeable battery, it is not limited but the various configuration members currently used conventionally can be used.
[0019] For example, a compound metallic oxide with the lithium which contains cobalt or nickel as positive active material is used. Only one kind may be chosen and used for such positive active material, and may be used for it combining two or more kinds. As such a compound metallic oxide, LiCoO2, LiNiO2, LiCo1-xNixO2 (0.01< x<1), etc. are mentioned, for example. Moreover, you may use it for LiCoO2, and LiMn 2O4, LiCoO2 and LiNiO2 and LiMn2O4, mixing suitably like LiNiO2.
[0020] a positive electrode -- the aforementioned positive active material -- binders, such as electric conduction agents, such as acetylene black and carbon black, polytetrafluoroethylene (PTFE), and polyvinylidene fluoride (PVDF), and a solvent -- kneading -- a positive electrode -- after considering as a mixture, this positive-electrode ingredient is applied to the lath plate of the aluminium foil as a charge collector, or the product made from stainless steel, and it is produced after desiccation and pressurization molding by heat-treating under a vacuum at the temperature of 50 degrees C - about 250 degrees C for about 2 hours.
[0021] Matter, such as a carbon material [the pyrolytic carbon, the corks, the graphite, the organic high-molecular-compounds (artificial-graphite, natural graphite, etc.) combustion object, and the carbon fiber], a compound stannic-acid ghost, etc. which have the graphite mold crystal structure which can emit [occlusion and] a lithium metal, a lithium alloy, and a lithium as a negative-electrode active material, is used. It is desirable to use the carbon material which has especially the graphite mold crystal structure whose spacing (d002) of a lattice plane (002) is 0.335-0.340nm. Only one kind may be chosen and used for these negative-electrode active materials, and may be used for them combining two or more kinds. in addition, a powder ingredient like a carbon material -- binders, such as an ethylene-propylene-diene terpolymer (EPDM), polytetrafluoroethylene (PTFE), and polyvinylidene fluoride (PVDF), -- kneading -- a negative electrode -- it is used as a mixture. Especially the manufacture approach of a negative electrode is not limited, but can be manufactured by the manufacture approach of the above-mentioned positive electrode, and the same approach.
[0022] Especially the structure of a lithium secondary battery is not limited and the coin mold cell which has the separator of a positive electrode, a negative electrode and a monolayer, or a double layer, a cylindrical cell, a square shape cell which has the separator of the shape of a positive electrode, a negative electrode, and a roll further, etc. are mentioned as an example. In addition, the fine porosity film of polyolefine well-known as a separator, textile fabrics, a nonwoven fabric, etc. are used.
[0023]
[Example] Next, an example and the example of a comparison are given and this invention is explained concretely.
Example 1 [preparation of nonaqueous electrolyte]
After having prepared the non-aqueous solvent of EC:PC:DEC(capacity factor) =30:5:65, having dissolved so that it might become the concentration of 1M about LiPF6 at this, and preparing nonaqueous electrolyte, tert-pentyl benzene was further added so that it might become 2.0 % of the weight to nonaqueous electrolyte.
[0024] [Production of a lithium secondary battery and measurement of a cell property] What mixed acetylene black (electric conduction agent) 10% of the weight 80% of the weight, mixed polyvinylidene fluoride (binder) at 10% of the weight of a rate, added the 1-methyl-2-pyrrolidone solvent to this, and was mixed was applied on aluminium foil, it dried, LiCoO2 (positive active material) was pressurization-cast, it heat-treated, and the positive electrode was prepared. It dried and pressurization-cast, applied what mixed the artificial graphite (negative-electrode active material) 90% of the weight, mixed polyvinylidene fluoride (binder) at 10% of the weight of a rate, added the 1-methyl-2-pyrrolidone solvent to this, and was mixed on copper foil and heat-treated, and the negative electrode was prepared.

And using the separator of a polypropylene fine porosity film, the above-mentioned nonaqueous electrolyte was made to pour in and the coin cell (3.2mm in the diameter of 20mm, thickness) was produced. Using this coin cell, under the room temperature (20 degrees C), it charged to termination electrical-potential-difference 4.2V for 5 hours, and then discharged to termination electrical-potential-difference 2.7V under 0.8mA constant current, and this charge and discharge were repeated by 0.8mA constant current and the constant voltage. Initial charge-and-discharge capacity is 1M which do not add a tert-alkylbenzene derivative. It was almost equivalent to the case (example 1 of a comparison) where LiPF6-EC/PC/DEC (capacity factor 30/5/65) is used as nonaqueous electrolyte, and when the cell property after 50 cycles was measured, the discharge capacity maintenance factor when making initial discharge capacity into 100% was 92.8%. Moreover, the low-temperature property was also good. The production conditions and cell property of a coin cell are shown in Table 1.

[0025] When used tert-pentyl benzene 5.0% of the weight to nonaqueous electrolyte as example 2 additive, and also nonaqueous electrolyte was prepared like the example 1, the coin cell was produced and the cell property after 50 cycles was measured, the discharge capacity maintenance factor was 91.5%. The production conditions and cell property of a coin cell are shown in Table 1.

[0026] When used tert-pentyl benzene 0.5% of the weight to nonaqueous electrolyte as example 3 additive, and also nonaqueous electrolyte was prepared like the example 1, the coin cell was produced and the cell property after 50 cycles was measured, the discharge capacity maintenance factor was 90.3%. The production conditions and cell property of a coin cell are shown in Table 1.

[0027] The non-aqueous solvent of example of comparison 1 EC:PC:DEC(capacity factor) =30:5:65 was prepared, and it dissolved so that it might become the concentration of 1M about LiPF6 at this. At this time, the tert-alkylbenzene derivative was not added at all. The coin cell was produced like the example 1 using this nonaqueous electrolyte, and the cell property was measured. The discharge capacity maintenance factor after 50 cycles was 82.6% to initial discharge capacity. The production conditions and cell property of a coin cell are shown in Table 1.

[0028] after preparing the non-aqueous solvent of example 4 EC:PC:DEC(capacity factor) =30:5:65, dissolving so that it may become the concentration of 1M about LiPF6 at this, and adjusting nonaqueous electrolyte -- further -- 4-tert-pentyl toluene was added so that it might become 2.0 % of the weight to nonaqueous electrolyte. The place which produced the coin cell like the example 1 using this nonaqueous electrolyte, and measured the cell property, Initial discharge capacity is tert-alkylbenzene derivative additive-free 1M. Are almost equivalent to the case (example 1 of a comparison) where LiPF6-EC/PC/DEC (capacity factor 30/5/65) is used as nonaqueous electrolyte. When the cell property after 50 cycles was measured, the discharge capacity maintenance factor when making initial discharge capacity into 100% was 92.1%. Moreover, the low-temperature property was also good. The production conditions and cell property of a coin cell are shown in Table 1.

[0029] When used benzene (1 and 1-diethyl propyl) 2.0% of the weight to nonaqueous electrolyte as example 5 additive, and also nonaqueous electrolyte was prepared like the example 1, the coin cell was produced and the cell property after 50 cycles was measured, the discharge capacity maintenance factor was 91.9%. The production conditions and cell property of a coin cell are shown in Table 1.

[0030] When used EC/PC/DEC/DMC (capacity factor 30/5/30/35), and replaced with the artificial graphite as a negative-electrode active material, and used the natural graphite, and also nonaqueous electrolyte was prepared like the example 1 as example 6 non-aqueous solvent, the coin cell was produced and the cell property after 50 cycles was measured, the discharge capacity maintenance factor was 92.8%. The production conditions and cell property of a coin cell are shown in Table 1.

[0031] As example 7 nonaqueous electrolyte, it is 1M. When used LiPF6-EC/PC/MEC/DMC (capacity factor 30/5/50/15), replaced with LiCoO2, and used LiNi0.8CO 0.2O2 as positive active material, and also nonaqueous electrolyte was prepared like the example 1, the coin cell was produced and the cell property after 50 cycles was measured, the discharge capacity maintenance factor was 91.1%. The production conditions and cell property of a coin cell are shown in Table 1.

[0032] As example 8 nonaqueous electrolyte, it is 1M. When used LiBF4-EC/PC/DEC/DMC (capacity factor 30/5/30/35), replaced with LiCoO2, and used LiMn 2O4 as positive active material, and also

nonaqueous electrolyte was prepared like the example 1, the coin cell was produced and the cell property after 50 cycles was measured, the discharge capacity maintenance factor was 92.6%. The production conditions and cell property of a coin cell are shown in Table 1.

[0033] When used 4-fluoro-tert-pentyl benzene 3.0% of the weight to nonaqueous electrolyte as example 9 additive, and also nonaqueous electrolyte was prepared like the example 1, the coin cell was produced and the cell property after 50 cycles was measured, the discharge capacity maintenance factor was 92.7%. The production conditions and cell property of a coin cell are shown in Table 1.

[0034] When used toluene 3.0% of the weight to nonaqueous electrolyte as example of comparison 2 additive, and also nonaqueous electrolyte was prepared like the example 1 of a comparison, the coin cell was produced and the cell property after 50 cycles was measured, the discharge capacity maintenance factor was 81.3%. The production conditions and cell property of a coin cell are shown in Table 1.

[0035] When used n-butylbenzene 3.0% of the weight to nonaqueous electrolyte as example of comparison 3 additive, and also nonaqueous electrolyte was prepared like the example 1 of a comparison, the coin cell was produced and the cell property after 50 cycles was measured, the discharge capacity maintenance factor was 79.7%. The production conditions and cell property of a coin cell are shown in Table 1.

[0036] When used di-n-butyl phthalate 3.0% of the weight to nonaqueous electrolyte as example of comparison 4 additive, and also nonaqueous electrolyte was prepared like the example 1 of a comparison, the coin cell was produced and the cell property after 50 cycles was measured, the discharge capacity maintenance factor was 78.1%. The production conditions and cell property of a coin cell are shown in Table 1.

[0037] When used 4-fluoro toluene 3.0% of the weight to nonaqueous electrolyte as example of comparison 5 additive, and also nonaqueous electrolyte was prepared like the example 1 of a comparison, the coin cell was produced and the cell property after 50 cycles was measured, the discharge capacity maintenance factor was 80.6%. The production conditions and cell property of a coin cell are shown in Table 1.

[0038]

[Table 1]

	正極	負極	化合物	添加量 wt%	電解液組成 (容量比)	初期放電容量 (相対値)	50サイクル放電容量維持率%
実施例1	$LiCoO_2$	人造黒鉛	tert-ペンチルベンゼン	2.0	1M $LiPF_6$ EC/PC/DEC=30/5/65	1.03	92.8
実施例2	$LiCoO_2$	人造黒鉛	tert-ペンチルベンゼン	5.0	1M $LiPF_6$ EC/PC/DEC=30/5/65	1.02	91.5
実施例3	$LiCoO_2$	人造黒鉛	tert-ペンチルベンゼン	0.5	1M $LiPF_6$ EC/PC/DEC=30/5/65	1.01	90.3
比較例1	$LiCoO_2$	人造黒鉛	なし	0	1M $LiPF_6$ EC/PC/DEC=30/5/65	1.00	82.6
実施例4	$LiCoO_2$	人造黒鉛	1-メチル-4-tert-ペンチルベンゼン	2.0	1M $LiPF_6$ EC/PC/DEC=30/5/65	1.02	92.1
実施例5	$LiCoO_2$	人造黒鉛	(1,1-ジエチルプロピル)ベンゼン	2.0	1M $LiPF_6$ EC/PC/DEC=30/5/65	1.02	91.9
実施例6	$LiCoO_2$	天然黒鉛	tert-ペンチルベンゼン	2.0	1M $LiPF_6$ EC/PC/DEC/DMC =30/5/30/35	1.02	92.8
実施例7	$LiNi_{0.8}Co_{0.2}O_2$	人造黒鉛	tert-ペンチルベンゼン	2.0	1M $LiPF_6$ EC/PC/MEC/DMC =30/5/50/15	1.15	91.1
実施例8	$LiMn_2O_4$	人造黒鉛	tert-ペンチルベンゼン	2.0	1M $LiBF_4$ EC/PC/DEC/DMC =30/5/30/35	0.99	92.6
実施例9	$LiCoO_2$	人造黒鉛	4-フルオロ-tert-ペンチルベンゼン	3.0	1M $LiPF_6$ EC/PC/DEC=30/5/65	1.02	92.7
比較例2	$LiCoO_2$	人造黒鉛	トルエン	3.0	1M $LiPF_6$ EC/PC/DEC=30/5/65	0.98	81.3
比較例3	$LiCoO_2$	人造黒鉛	n-ブチルベンゼン	3.0	1M $LiPF_6$ EC/PC/DEC=30/5/65	0.97	79.7
比較例4	$LiCoO_2$	人造黒鉛	ジ-n-ブチルフタレート	3.0	1M $LiPF_6$ EC/PC/DEC=30/5/65	0.97	78.1
比較例5	$LiCoO_2$	人造黒鉛	4-フルオロトルエン	3.0	1M $LiPF_6$ EC/PC/DEC=30/5/65	0.98	80.6

[0039] In addition, this invention is not limited to the example of a publication, but various combination which can be guessed is easily possible for it from the meaning of invention. Especially the combination of the solvent of the above-mentioned example is not limited. Furthermore, although the above-mentioned example is related with a coin cell, this invention is applied also to the cell of a cylindrical shape and a prism form.

[0040]

[Effect of the Invention] According to this invention, the lithium secondary battery excellent in cell properties, such as the cycle property of a cell, electric capacity, and a preservation property, can be offered.

[Translation done.]